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Green synthesis of ZSM-5 using biomass fly ash and recycled mother liqour

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Abstract

The effects of synthesis conditions, silica derived from biomass fly ash, and mother liquor recycling on the synthesis of ZSM-5 were investigated using both hydrothermal and clear solution methods. The synthesis of ZSM-5 from TEOS was optimized using a hydrothermal autoclave reactor and reflux assisted by microwave treatment, revealing that the combined reflux–microwave approach significantly enhances crystallization, resulting in high-quality ZSM-5. In contrast, reduced synthesis time and milder conditions (e.g., lower pressure) proved ineffective, highlighting the need for more rigorous conditions to promote zeolite formation. For biomass fly ash-derived silica, two distinct molar ratios were tested, demonstrating that the ratio 25Si:1Al:9TPAOH:0.16NaOH:495H2O:100EtOH was more effective than $1Al_2O_3$:$40SiO_2$:0.2TPAOH, which produced a mixture of *silicalite-1* and amorphous phases. Recycling the mother liquor from ZSM-5 synthesis yielded comparable product quantities and preserved well-defined crystallinity, although excess sodium ions reduced yields. Scanning electron microscopy showed that recycling resulted in smaller, well-dispersed crystals, suggesting enhanced nucleation and crystal growth. Overall, this study highlights the critical role of synthesis conditions and precursor selection in the efficient, resource-conscious production of ZSM-5 from biomass waste.

Keywords: Biomass fly ash, Green synthesis, Mother liqour recycling, ZSM-5

1. Introduction

ZSM-5 is a zeolite characterized by its distinctive 10-membered ring structure, comprising both straight and sinusoidal channels that are essential for catalysis and separation processes [1]. Its framework is composed of silicon and aluminum atoms linked by oxygen atoms, forming the Mobil Five (MFI) structure [2]. The acidic properties of ZSM-5 enhance its catalytic performance, making it suitable for a wide range of organic transformations [3, 4]. This zeolite is widely utilized in petroleum refining, organic synthesis, and environmental applications, including air and water purification.

The use of commercial silica sources such as tetraethyl orthosilicate (TEOS), together with organic templates like tetrapropylammonium hydroxide (TPAOH), offers advantages in zeolite synthesis but also presents economic and environmental challenges when compared with more cost-effective alternatives [5, 6]. While TEOS and TPAOH enable precise control over synthesis parameters, their relatively high cost significantly increases production expenses, particularly at industrial scales [7]. TEOS requires stringent control of hydrolysis and condensation reactions [8], adding complexity and necessitating specialized equipment. Furthermore, TPAOH's high alkalinity demands careful handling due to associated environmental and safety concerns. Both TEOS and TPAOH may pose environmental risks through potential chemical emissions during synthesis [6].

To address these issues, alternative silica sources such as fly ash and more economical templates have been explored to reduce production costs and environmental impact. Biomass fly ash, a byproduct of biomass-based power generation, is increasingly available due to the growing demand for renewable energy to replace fossil fuels [9]. This trend has led to increased interest in valorizing biomass fly ash as an industrial feedstock. For example, biomass electricity generation in Thailand rose from 1,959.95–2,451.82 MW during 2012–2014 to 3,851.51 and

3,869.81 MW in 2022 and 2023, respectively. The national target for 2037 is 5,790.00 MW [10, 11]. Biomass fly ash is typically rich in silica and contains minimal contaminants, particularly heavy metals, making it a promising silica source for zeolite synthesis.

In addition to using alternative feedstocks, recycling the mother liquor—containing residual reagents—can further reduce dependence on commercial silica and organic templates [6]. TPAOH is widely applied in diverse fields, including as a surfactant, in semiconductor manufacturing, and in aqueous rechargeable batteries. TEOS is commonly used in the semiconductor industry for silicon dioxide film deposition, offering a safer alternative to pyrophoric silane. Recycling mother liquor not only enhances sustainability by recovering reagents for reuse across industries but also contributes to environmental conservation.

This study investigates a green synthesis route for ZSM-5 using silica extracted from biomass fly ash and recycled mother liquor, aiming to reduce production costs and valorize industrial byproducts. It evaluates the suitability of fly ash as an underutilized silica source and assesses the feasibility of precursor recycling. The influence of molar ratios, synthesis methods, and conditions on ZSM-5 formation is also systematically examined.

2. Materials and methods

2.1 Extraction of Silica from Biomass Fly Ash

Biomass fly ash was obtained from a power plant located in eastern Thailand that uses biomass as fuel. The sample was dried at 70 °C for 1 hour and stored in a moisture-absorbing container. For silica extraction, the fly ash was hydrothermally treated with 1 M NaOH at 100 °C for 24 hours. The mixture was then filtered, and the solid residue was rinsed with hot deionized water. Silica was precipitated by adjusting the pH of the filtrate to 7.0 using an acetic acid solution. After aging for 24 hours, the precipitate was centrifuged at 2,500 rpm and washed twice to remove soluble salts. The silica was subsequently oven-dried at 80 °C for 24 hours. X-ray analysis (Horiba XGT-2000W) confirmed a high silica purity of 99.42% w/w [12].

2.2 ZSM-5 Synthesis

ZSM-5 was synthesized using silica sourced from either tetraethyl orthosilicate (TEOS, Aldrich) or biomass fly ash, along with ≥98% aluminum isopropoxide (AIP, Aldrich) and a 40 wt% aqueous solution of TPAOH, Merck, following a modified method based on Song et al. [13].

Biomass fly ash-derived silica was dissolved in either 0.1 M or 0.001 M NaOH, corresponding to molar ratios of $9TPAOH:0.16NaOH:1Al:25Si:495H_2O:100EtOH$ and $1Al_2O_3:40SiO_2:0.2TPAOH$, respectively. All chemicals used were of analytical grade. The synthesis gel was prepared by mixing AIP, TPAOH, either TEOS or biomass-derived silica, NaOH, and deionized water.

The mixture was stirred at room temperature until a clear solution was obtained. The solution was then transferred to a hydrothermal autoclave reactor equipped with a polytetrafluoroethylene (PTFE) chamber and heated under the specified conditions. After synthesis, crystals were recovered via multiple centrifugation cycles at 3,000 rpm and washed with deionized water. The mother liquor (i.e., the supernatant from the first centrifugation cycle) was reused in subsequent synthesis. This process was repeated for the following cycles. The recovered solids were dried at 110 °C and calcined at 550 °C to remove the TPAOH template. Product yield was calculated by dividing the weight of the final product by the weight of silica used.

2.3 Characterization of Synthesized Crystals

The synthesized crystals were characterized by X-ray diffraction (XRD) using a Malvern Panalytical Empyrean instrument. XRD measurements were carried out over a 2θ range of 5°–45° to identify the crystalline phases of the zeolite. Surface morphology, particle shape, and size were examined by scanning electron microscopy (SEM) using a Philips XL30 instrument.

3. Results and discussion

3.1 ZSM-5 Synthesis from Commercial Silica Source (TEOS)

ZSM-5 synthesis using commercial silica derived from TEOS was performed under a range of experimental conditions specific to this study (Table 1). Based on the methodology of Song et al. [13], which uses a molar ratio of $25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$ at 165 °C for 120 h, effective synthesis of ZSM-5 was achieved. This process was conducted using either a hydrothermal autoclave reactor equipped with a PTFE chamber or a reflux system combined with microwave irradiation.

After synthesis, the initially clear solution became opaque and milky, forming a colloidal suspension (Figure 1). XRD analysis confirmed the formation of ZSM-5 crystals under both synthesis conditions (Figure 2). The selected precursor ratios were optimal, and the combination of elevated temperature and extended reaction duration effectively promoted zeolite nucleation. Furthermore, the use of a pressure reactor or microwave-assisted reflux significantly enhanced both crystallization and crystal growth. These results align with the findings of [14], who reported that prolonged microwave irradiation and higher energy levels facilitate zeolite A crystallization by accelerating the dissolution of Al and Si from coal fly ash.

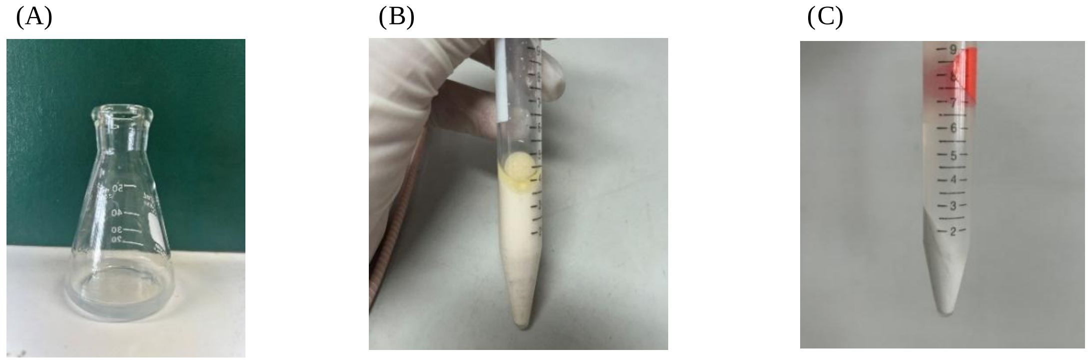

Figure 1 Observations of the mixtures at some stages of zeolite synthesis: (A) after mixing (clear solution), (B) after heating (milk-like solution) and (C) after washing synthesized products.

Table 1 Initial compositions, synthesis parameters and properties of the synthesized products.

Expt		Composition	Mother liquid recycling (cycle)	Temp (ºC)	Time (h)	Method	Product yield (% of silica wt.)	Product phase (XRD)
		Silica from TEOS[†]						
1		$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	-	165	120	Reflux[‡]	-	Clear solution
2		$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	-	165	120	Microwave	-	Clear solution
3	a	$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	-	165	120	Reflux (+microwave) *	N.A.*	ZSM-5
4	b	$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	-	165	120	Pressure reactor***	N.A.	ZSM-5
5		$25Si:1NaAlO_2:9TPAOH:0.16NaOH:300H_2O$	-	165	72	Reflux	-	Clear solution
6		SiO_2/Al_2O_3 = 80, 0.0768g $NaAlO_2$, 0.5 ml TPAOH, 424 mg NaOH, 49.2 ml H_2O	-	150	24	Reflux	-	Clear solution
7		SiO_2/Al_2O_3 = 80, 0.0768g $NaAlO_2$, 0.5 ml TPAOH, 424 mg NaOH, 49.2 ml H_2O	-	150	48	Reflux	-	Clear solution
		Silica from biomass fly ash						
8	c	$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	1st batch	165	120	Pressure reactor	34.89	ZSM-5
9	d	$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	2nd batch (1st cycle)	165	120	Pressure reactor	30.83	ZSM-5
10	e	$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	3rd batch (2nd cycle)	165	120	Pressure reactor	35.82	ZSM-5
11		$25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH$	4th batch (3rd cycle)	165	120	Pressure reactor	28.89	N.A.
12	f	$1Al_2O_3:40SiO_2:0.2TPAOH$	1st batch	210	2	Pressure reactor	56.21	*Silicalite-1* + amorphous
13	g	$1Al_2O_3:40SiO_2:0.2TPAOH$	2nd batch (1st cycle)	210	2	Pressure reactor	-	Amorphous
14	h	$1Al_2O_3:40SiO_2:0.2TPAOH$	3rd batch (2nd cycle)	210	2	Pressure reactor	-	Amorphous

Description† TEOS = commercial silica; ‡ Reflux and microwave treatment at 80 ºC for 30 min and then 165 ºC for 1 h; *N.A. = not analyzed; ***Autoclave pressure reactor

Samples treated via reflux and microwave exhibited sharper ZSM-5 XRD peaks compared to the broader peaks obtained from the autoclave reactor (Figure 2). Broad peaks, although characteristic of ZSM-5, may reflect structural irregularities or reduced crystal size. Harsh synthesis conditions, such as elevated temperature and pressure, can affect structural uniformity, while the more moderate conditions of the reflux method may have allowed a slower, more orderly crystallization process. Consequently, XRD patterns suggest that smaller crystal sizes were achieved under reflux and microwave conditions relative to those formed in the autoclave reactor.

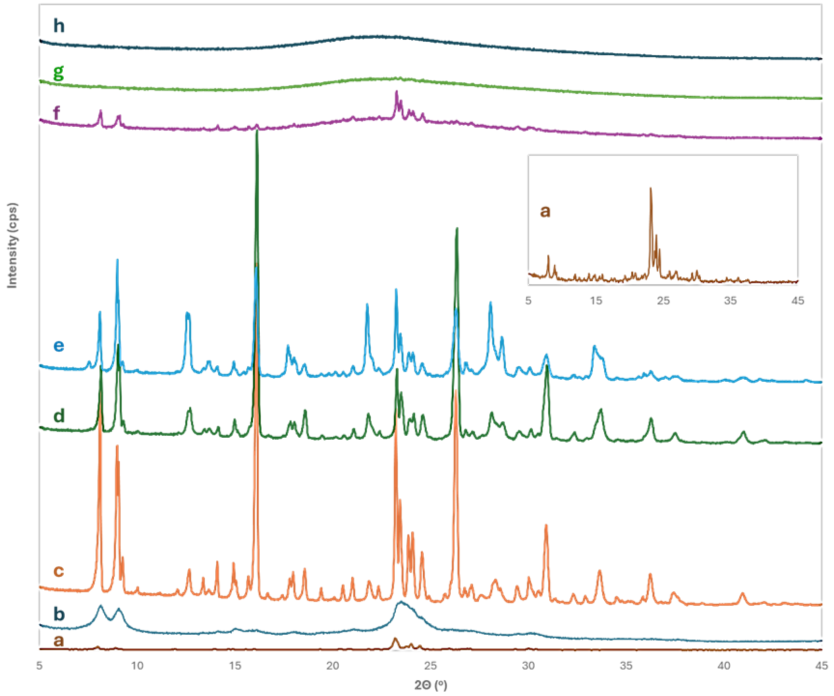

Figure 2 XRD profiles of synthesized samples under various experimental conditions (see Table 1). The X and Y axes represent 2θ and intensity (cps), respectively. (a) 25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH (silica from TEOS), reflux + microwave; (b) same as (a), synthesized in a pressure reactor; (c–e) as (b), using silica from biomass fly ash, for the 1st to 3rd batches; (f–h) 1Al_2O_3:40SiO_2:0.2TPAOH (silica from biomass fly ash), for the 1st to 3rd batches.

Importantly, the synthesis method in Song et al. [13], which used reflux and microwave treatment independently, resulted in a clear solution or gel, indicating the absence of ZSM-5 formation (Table 1). Several factors may explain this: (1) the reflux system operates at atmospheric pressure, which supports nucleation but lacks sufficient energy for crystal growth. For successful ZSM-5 synthesis via hydrothermal treatment, high pressure (>1 bar) and temperature (>373 K) are essential [15]; (2) microwave treatment alone does not provide the specific conditions and duration needed to promote nucleation and growth of ZSM-5. Wilayat and Annisa [16] also found that longer crystallization times for ZSM-5 lead to higher catalyst crystallinity; and (3) combining reflux and microwave heating with a pressure reactor yields optimal conditions for nucleation, crystallization, and crystal growth in this study.

To investigate synthesis time reduction, a molar ratio of 25Si:1$NaAlO_2$:9TPAOH:0.16NaOH:300H_2O was tested under conditions of 165 °C for 72 h. The resulting solution remained clear, and no ZSM-5 crystals were observed; thus, XRD analysis was not performed (Table 1). The lower H_2O ratio was intended to enhance molecular interaction and promote crystallization. However, this adjustment did not yield crystal formation, likely due to the shortened synthesis time and insufficient reaction intensity.

Zeolite synthesis is often limited by the prolonged high-temperature conditions required to dissolve silicon and aluminum sources. These conditions can hinder the formation of specific zeolite frameworks, especially those with high porosity [14]. To address this, molar ratios proposed by Panpa and Jinawath [5] were tested using the reflux method for 24 and 48 h (Table 1). No ZSM-5 crystals were detected, as the solution remained clear even after 48 h. By contrast, Panpa and Jinawath [5] achieved ZSM-5 crystallization within 24 h under similar conditions using a stainless-steel bomb reactor at autogenous pressure. They found that SiO_2/Al_2O_3 ratios between 80 and 2075 led to well-crystallized ZSM-5, whereas ratios below 80 produced poorly crystalline or amorphous products. These discrepancies underscore the critical role of both sufficient reaction intensity and duration in promoting successful ZSM-5 nucleation and growth.

3.2 ZSM-5 Synthesis by Using Silica from Biomass Fly Ash

3.2.1 Synthesis Conditions

ZSM-5 synthesis using silica extracted from biomass fly ash was investigated under two distinct molar compositions: 25Si:1Al:9TPAOH:0.16NaOH:495H_2 O:100EtOH (as reported by Song et al. [13]) and 1Al_2O_3:40SiO_2:0.2TPAOH (modified from Hanyotee and Chareonpanich [17]). XRD profiles of the synthesized products are presented in Table 1 and Figure 1. The results indicate that the composition proposed by Song et al. [13] was more effective for producing ZSM-5 using biomass fly ash-derived silica. In contrast, the method adapted from Hanyotee and Chareonpanich [17] yielded a mixture of *silicalite-1*—a pure silica polymorph of ZSM-5 [18]—and amorphous phases.

The SiO_2/Al_2O_3 molar ratio of 40 used in the Hanyotee and Chareonpanich [17] method, which is higher than the ratio of 25 used in Song et al. [13]. likely contributed to the formation of *silicalite-1*. This suggests that the synthesis conditions under the higher SiO_2/Al_2O_3 ratio were insufficient for full transformation into ZSM-5. Furthermore, while Hanyotee and Chareonpanich [17] used tetra-n-propylammonium bromide (TPABr) as the structure-directing agent, this study used TPAOH. TPABr, a quaternary ammonium salt with low water solubility, is particularly effective under acidic conditions and facilitates faster ZSM-5 crystallization due to its lower activation energy [19]. In contrast, TPAOH is highly water-soluble and imparts basicity due to hydroxide ions. The use of TPAOH under the synthesis conditions adapted from Hanyotee and Chareonpanich [17] may have resulted in an overly basic environment, hindering complete ZSM-5 crystallization.

To minimize the formation of amorphous phases in future syntheses, strategies such as optimizing the SiO_2/Al_2O_3 ratio, fine-tuning synthesis parameters (e.g., temperature, duration, and base concentration), and adding stabilizing agents should be considered. Additionally, ZSM-5 seeding or the reuse of recycled mother liquor from successful syntheses may enhance nucleation and improve crystallinity.

3.2.2 Mother Liquor Recycling

The mother liquor remaining after ZSM-5 crystallization contains residual reagents, including unreacted silica and alumina, hydroxide ions, organic templates (e.g., TPAOH), and zeolite nuclei. Reusing this solution in subsequent syntheses enhances resource efficiency. In this study, mother liquor recycling was performed following the approach of Song et al. [13], yielding ZSM-5 with product yields ranging from 28.9% to 35.8%, based on the mass of silica used. XRD analysis confirmed the successful formation of ZSM-5 in both the first and second recycling cycles. The resulting XRD patterns displayed sharp and distinct peaks, indicating the retention of well-defined ZSM-5 crystallinity (Table 1). These findings are consistent with Pan et al. [6], who observed no secondary phases in samples synthesized using recycled mother liquor.

However, yields from the recycled mother liquor were slightly lower than or comparable to those achieved with deionized (DI) water. This reduction is likely due to high NaOH concentrations, which can negatively affect crystallinity [18]. Hydroxide ions act as mineralizers, promoting the depolymerization of silica and polymerization of silicate and aluminate anions to accelerate zeolite growth. Sodium ions, as inorganic structure-directing agents, facilitate ZSM-5 crystallization. However, excess Na^+ disrupts the charge balance required to stabilize the framework, increasing the silicate concentration to compensate, and ultimately reducing zeolite yield [6]. Additionally, Yu, Kwon [20] reported that the ratio of tetramethylammonium hydroxide (TMAOH) to NaOH significantly influences crystal size, with higher ratios yielding smaller crystals—highlighting Na^+'s role in controlling crystal morphology.

In contrast, when using recycled mother liquor from a synthesis based on 1Al_2O_3:40SiO_2:0.2TPAOH (adapted from [17]), no zeolite formation was observed. This outcome can be attributed to the inappropriate precursor ratios and the choice of template. The ionic strength of the recycled solution may have further hindered nucleation, especially when *silicalite-1* formed during initial synthesis. The presence of *silicalite-1* seeds in the recycled mother liquor likely promoted competing nucleation pathways, unfavourable for ZSM-5 formation. Consequently, both first and second reuse cycles resulted in amorphous silica precipitation under the experimental conditions.

3.2.3 SEM Analysis

SEM images of the synthesized samples are presented in Figure 3. For the molar ratio 25Si:1Al:9TPAOH:0. 16NaOH:495H2O:100EtOH, the first synthesis batch exhibited crystal agglomeration, consisting of larger hexagonal particles interspersed with smaller, clustered crystals. In contrast, the use of recycled mother liquor resulted in more dispersed ZSM-5 crystals with slightly reduced particle sizes. This suggests that recycling enhances nucleation by reintroducing zeolite nuclei, leading to more uniform and smaller crystals in subsequent batches. These observations are consistent with Pan et al. [6], who also reported smaller ZSM-5 crystals formed with recycled mother liquor due to increased nucleation sites.

For the synthesis based on 1Al2O3:40SiO2:0.2TPAOH [17], a mixture of silicalite-1 and amorphous materials was observed, displaying diverse particle morphologies, including rectangular and spheroidal shapes (Figure 2). The spheroidal forms are consistent with silicalite-1 synthesized using similar conditions, as described by Pengthamkeerati et al. [9].

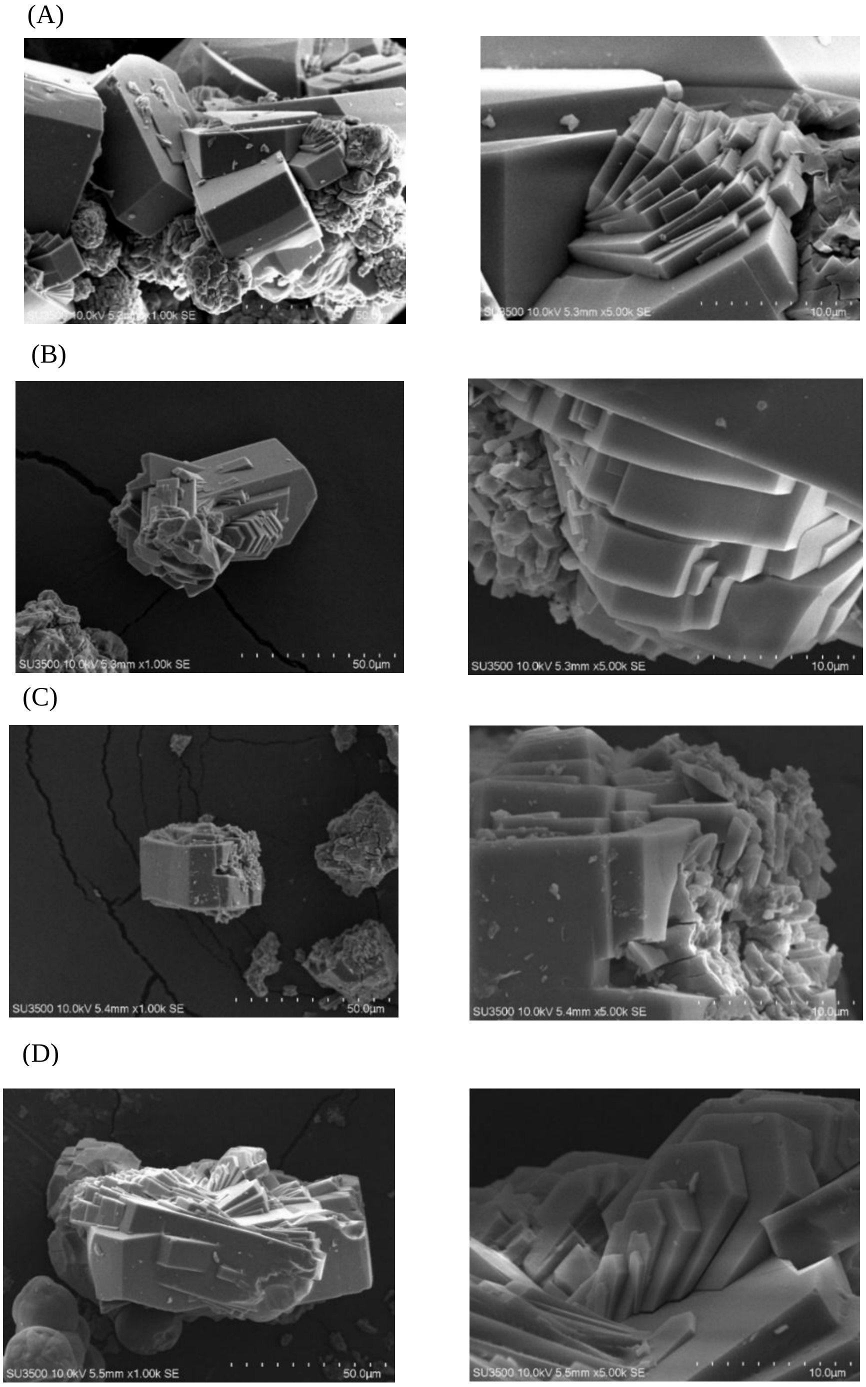

Figure 3 SEM images of the synthesized samples under different experimental conditions (see Table 1). The left panel shows a scale bar of 50 µm; the right panel shows a scale bar of 10 µm. (A) 25Si:1Al:9TPAOH: 0.16NaOH:495H2O:100EtOH (1st batch), (B) 25Si:1Al:9TPAOH:0.16NaOH:495H2O:100EtOH (2nd batch; 1st mother liquid recycling), (C) 25Si:1Al:9TPAOH:0.16NaOH:495H2O:100EtOH (3rd batch; 2nd mother liquid recycling), (D) 25Si:Al:9TPAOH:0.16NaOH:495H2O:100EtOH (4th batch; 3rd mother liquid recycling), (E) 1Al2O3:40SiO2:0.2TPAOH (1st batch).

(E)

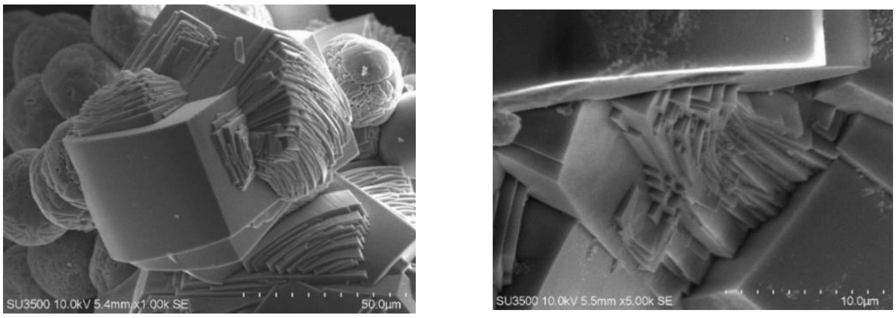

Figure 3 (cont.) SEM images of the synthesized samples under different experimental conditions (see Table 1). The left panel shows a scale bar of 50 µm; the right panel shows a scale bar of 10 µm. (A) 25Si:1Al: 9TPAOH: 0.16NaOH:495H2O:100EtOH (1st batch), (B) 25Si:1Al:9TPAOH:0.16NaOH:495H2O:100EtOH (2nd batch; 1st mother liquid recycling), (C) 25Si:1Al:9TPAOH:0.16NaOH:495H2O:100EtOH (3rd batch; 2nd mother liquid recycling), (D) 25Si:Al:9TPAOH:0.16NaOH:495H2O:100EtOH (4th batch; 3rd mother liquid recycling), (E) 1Al2O3:40SiO2:0.2TPAOH (1st batch).

4. Conclusions

This study demonstrated the successful synthesis of ZSM-5 using silica sourced from both commercial TEOS and biomass fly ash, emphasizing the critical influence of synthesis conditions and precursor materials. Using a molar ratio of 25Si:1Al:9TPAOH:0.16NaOH:495H_2O:100EtOH under the conditions established by Song et al. [13] enabled effective ZSM-5 crystallization. Hydrothermal autoclave treatment and reflux assisted by microwave activation were particularly effective in enhancing crystallization. In contrast, reduced synthesis times and milder conditions proved insufficient, reinforcing the necessity of vigorous reaction parameters for successful zeolite formation. When using silica derived from biomass fly ash, specific molar ratios and controlled conditions were essential for ZSM-5 production. The alternative ratio of 1Al_2O_3: 40SiO_2: 0.2TPAOH (based on Hanyotee and Chareonpanich [17]) resulted in a mixture of *silicalite-1* and amorphous phases, indicating incomplete zeolite transformation under those conditions. Moreover, recycling mother liquor from ZSM-5 synthesis proved to be a promising approach for improving resource efficiency. However, variations in ionic strength and elevated sodium ion concentrations negatively impacted product yield and crystallinity. Hence, this study confirms that ZSM-5 can be synthesized using biomass fly ash as a silica source, and that mother liquor recycling can contribute to a more sustainable and cost-effective production process, provided that synthesis parameters are appropriately controlled.

5. Acknowledgements

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6. Conflict of Interest

The authors declare that they have no conflict of interest.

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